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Study of Anion-Molecule Reaction Dynamics with Time-Dependent Photoelectron Spectroscopy

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FINAL TECHNICAL REPORT

TITLE: Study of Anion-Molecule Reaction Dynamics with Time-Dependent Photoelectron Spectroscopy

PRINCIPAL INVESTIGATOR: Daniel M. Neumark

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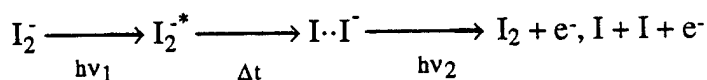
Abstract:

A novel femtosecond time-resolved experiment has been set up to probe the photodissociation dynamics of negative ions and negative ion clusters. First results for the photodissociation of I_2^- have been obtained recently.

Final Technical Report:

DURIP funds were used to purchase a Ti:sapphire femtosecond laser system (Model #CPA-1000 MPS) from Clark MRX Inc., at a cost of \$165,600. The Ti:sapphire femtosecond laser system is being used on a new experiment designed to study the photodissociation dynamics of negative ions and negative ion clusters on a femtosecond time scale. The novel feature of this experiment is that photoelectron spectroscopy is used to probe the dynamics of the dissociating anion in real-time. We have very recently obtained our first results on I_2^- photodissociation.

The principle of the experiment can be best understood by specifically considering its application to I_2^- :



This is a pump-probe experiment using two femtosecond laser pulses. The pump pulse ($h\nu_1$, 780 nm) excites mass-selected I_2^- from the ground $X^2\Sigma_u^+$ state to the repulsive $2\Pi_{u(3/2)}$ state. The I_2^- begins to dissociate, and after a variable time delay Δt , the dissociating ions are photodetached with the probe pulse ($h\nu_2$, 260 nm). We then measure the photoelectron spectrum, thereby mapping out the dissociating wave packet onto the well-known I_2 potential energy surfaces.

At short times, one expects to see a transient associated with the dissociating I_2^- molecule, and at longer times, the I^- photoelectron spectrum should become more prominent as dissociation occurs. This is precisely what has been observed in our first measurements on I_2^- . However, the time scale for dissociation appears to be several hundred fsec longer than predicted by our simulations that use the currently accepted potential energy curves for I_2^- , indicating that these potentials may require significant modification. This issue will be addressed in further experiments and theoretical work.